DIELECTRIC BARRIER DISCHARGES: PULSED BREAKDOWN, ELECTRICAL CHARACTERIZATION AND CHEMISTRY*

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Abstract

The application of atmospheric pressure discharges in new fields like environmental protection, surface treatment or life-sciences requires a profound knowledge on the plasma parameters and properties. This includes the characterization of the breakdown processes triggering plasma chemistry, the proper determination of the electrical parameters and the description of the dominant chemical pathways. This contribution aims to present new approaches regarding these three topics for pulsed driven Dielectric Barrier Discharges (DBDs) in particular. Fast electrical, optical and spectroscopic methods enable the study of ignition, breakdown statistics and spatio-temporally resolved development of pulsed DBD microdischarges. The determination of electrical parameters such as discharge current, gas gap voltage, instantaneous power and energy as well as the charge transferred through the gas gap is based on a simple equivalent circuit which is consistent with sinusoidal-voltage driven or miniature pulsed driven DBDs. The characterization of the dominant chemical pathways of advanced plasma processes discusses also several examples including secondary effects, such as adsorption-enhanced VOC conversion by DBD plasma treatment.

I. INTRODUCTION

More than 150 year ago an invention of the young engineer Werner Siemens (later Werner von Siemens) became the birth of non-thermal plasma chemical technology: the ozonizer. Nowadays non-thermal plasma technologies are state of the art for the generation of ozone as an important oxidant, for surface treatment and pollution degradation, in plasma displays and as light sources [1]. All these application use the same type of discharge, the Dielectric Barrier Discharge (DBD) also known as silent discharge, partial discharge or barrier discharge. Beside the aforementioned applications the use of DBDs for life-science issues is under study worldwide [2]. One of the fields of DBD application rapidly growing within the last years is exhaust gas treatment, in particular deodorization [3]. The application of atmospheric pressure discharges in all this fields requires a profound knowledge on the plasma parameters and properties. This includes

(1) the characterization of the breakdown processes triggering plasma chemistry,
(2) the proper determination of the electrical parameters and
(3) the description of the dominant chemical pathways.

This contribution aims to present new approaches regarding these three topics for pulsed driven DBDs in particular. It will shortly summarize new investigations on the ignition, breakdown and spatio-temporally resolved development of pulsed DBD microdischarges and the determination of electrical parameters based on a simple equivalent circuit.

II. GENERAL OVERVIEW

The following figure aims to summarize the different aspects of recent research activities in our laboratory. Any application oriented approach, in particular in case of pollution degradation in off-gases or exhaust must contain two issues, namely the fundamental knowledge about the plasma and the aspects of overall process development.

The challenge regarding plasma properties and parameters is that DBDs and other non-thermal plasmas at atmospheric pressure are filamentary gas discharges, i.e. they consists of many individual, short-lived and thin microdischarges forming filaments.

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14. ABSTRACT
The application of atmospheric pressure discharges in new fields like environmental protection, surface treatment or life-sciences requires a profound knowledge on the plasma parameters and properties. This includes the characterization of the breakdown processes triggering plasma chemistry, the proper determination of the electrical parameters and the description of the dominant chemical pathways. This contribution aims to present new approaches regarding these three topics for pulsed driven Dielectric Barrier Discharges (DBDs) in particular. Fast electrical, optical and spectroscopic methods enable the study of ignition, breakdown statistics and spatio-temporally resolved development of pulsed DBD microdischarges. The determination of electrical parameters such as discharge current, gas gap voltage, instantaneous power and energy as well as the charge transferred through the gas gap is based on a simple equivalent circuit which is consistent with sinusoidal-voltage driven or miniature pulsed driven DBDs. The characterization of the dominant chemical pathways of advanced plasma processes discusses also several examples including secondary effects, such as adsorption-enhanced VOC conversion by DBD plasma treatment.

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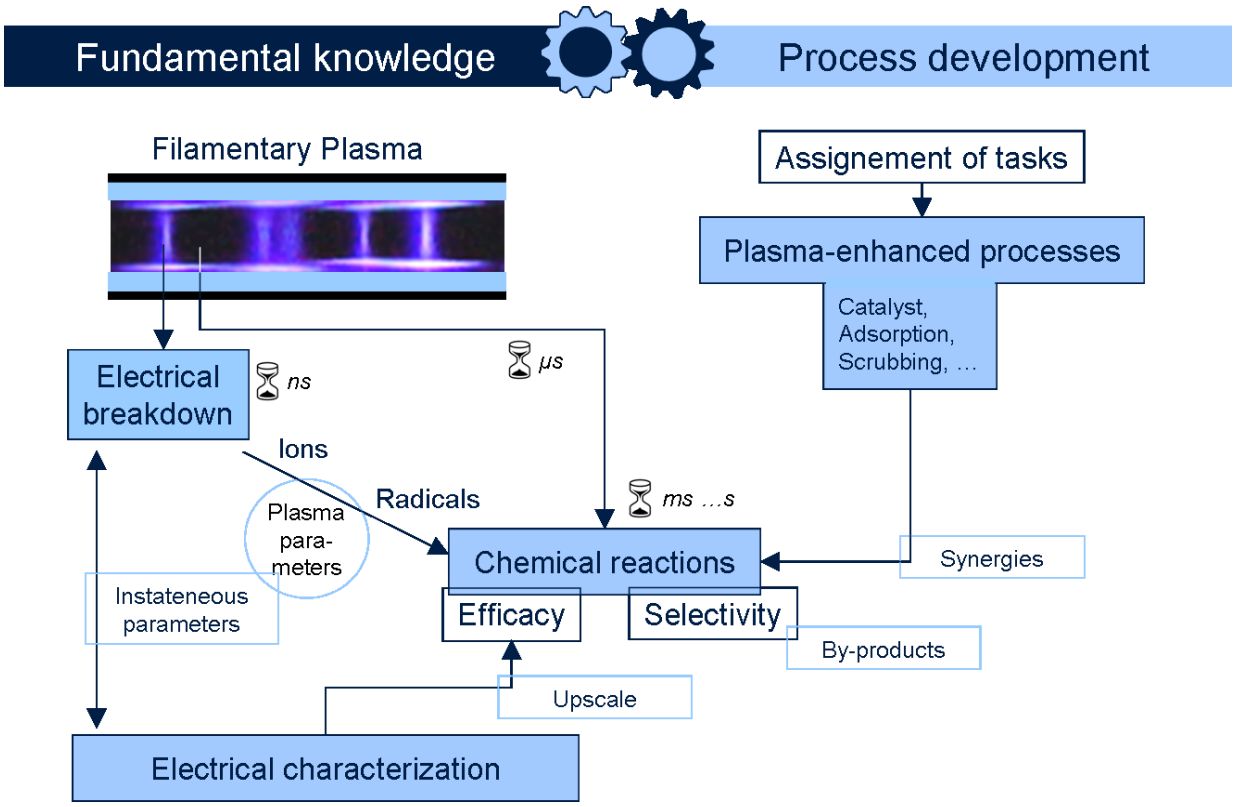

Figure 1. The different aspects of application oriented research on DBD-based pollution degradation.

The formation of a microdischarge proceeds within several ten nanoseconds while the desired chemical processes develop on time-scales in the order of micro- to milliseconds. Plasma-chemical conversion is via reactions including short-lived reactive species such as radicals and ions and thus determined by the plasma parameters (e.g. electron density, reduced electric field strength, electron energy distribution function). This requires the study of the microdischarges itself by means of fast electrical, optical and spectroscopic methods.

The target plasma-chemical conversion must be discussed under two aspects namely efficacy and selectivity. Efficacy means the energy cost to perform a certain number of chemical reactions, e.g. the energy yield to decompose a pollutant molecule into harmless products. Selectivity pays attention to the fraction of undesired by-products which can be generated in a plasma-chemical conversion. Non-thermal plasmas at moderate temperatures in oxygen containing gases (e.g. air with organic pollutants) are usually dominated by oxidation processes [1-4]. For example in exhaust gases the conversion of NO to NO_2 can be done with reasonable energy input [5] while the removal of NO_2 or higher oxides can be done by a catalytic processes or by means of scrubbing. Therefore, most commercial plasma processes in exhaust treatment applications are a combination of an oxidizing plasma stage with other technologies, e.g. catalyst, adsorbing agents or scrubbing. Such combinations are more than a serial connection of different processes and enable synergies which increase the efficacy and selectivity [6].

A profound investigation of the plasma properties, the efficacy of plasma chemistry and the upscale of laboratory results to real installations requires the electrical characterization, i.e. the determination of electrical parameters such as the discharge current, the gas gap voltage, the instantaneous power and energy as well as the charge transferred through the gas gap.

III. ELECTRICAL CHARACTERIZATION

The determination of electrical parameters is based on a equivalent circuit. Recently we reviewed different approaches carrying out that the circuit shown in fig. 2 is the simplest equivalent circuit suitable in most situations including the classical ozoniser and uniform DBDs in smale scale laboratory reactors [7]. The dielectric barrier is represented by the capacitance $\mathbf{C_d}$. This capacitance is in series with a parallel connection of a variable resistor **R(t)** and the gas gap capacity $\mathbf{C_g}$. This parallel connection is capable to cover the change of the effective capacitance and non-zero gas gap voltage $\mathbf{U_g(t)}$ during discharge ignition. This is particularly the case for pulsed driven DBDs when the applied voltage changes during the breakdown and the active plasma phase. Note that the simplification $C_g/C_d << 1$ is not necessarily valid, i.e. the

capacitance values must be determined in order to derive the exact values and slopes of the dissipated discharge current, instantaneous power and energy. Instead of C_g, the total capacity C_{cell} ($1/C_{cell} = 1/C_d + 1/C_g$) can be used. This is recommended since C_{cell} can be determined experimentally from voltage-chage-plots (often referred too as Lissajous-figures) [8]. Further details can be found in our recent papers [7-9].

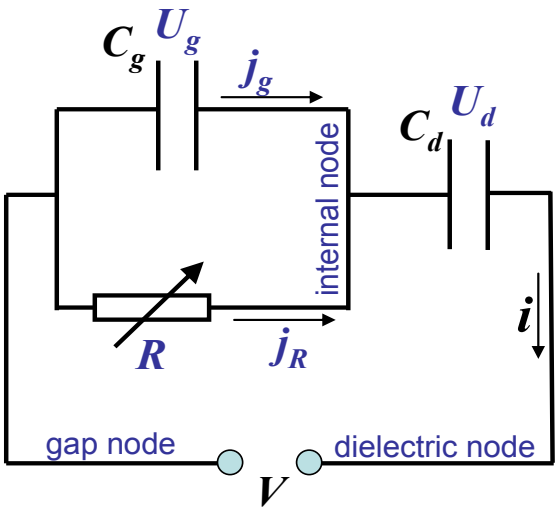

Figure 2. Equivalent circuit of a DBD arrangement.

The study of electrical parameters gives insights into the character of DBD, e.g. filamentary or diffuse discharges. Furthermore, it could be demonstrated that the energy dissipated per cycle can depend on applied voltage waveform [10]. Examples are shown in fig. 3.

The data in fig. 3 has been derived for localized microdischarges in a special symmetric DBD arrangement consisting of two semi-spherical electrodes covered by Al_2O_3 as barrier material.

In case of rectangular high voltage (switched DC-voltage with 10 kV amplitude) two microdischarges are observed, namely in the rising and the falling slope of the high voltage pulse. The bottom diagram in fig. 3 show that a larger discharge current amplitude as well as a longer discharge duration are obtained in case of pulsed DBD operation compared with a sinusoidal high voltage (14 kV amplitude). The time between rising and falling slope t_{pulse} could be changed and resulted in a significant change of the discharge current in the falling slope (top diagram in fig. 3). As shorter the time between the microdischarge in the rising slope and in the falling slope as lower is the discharge current amplitude but as longer is the discharge duration. This is related to a manipulation of the volume breakdown as discussed in the next section.

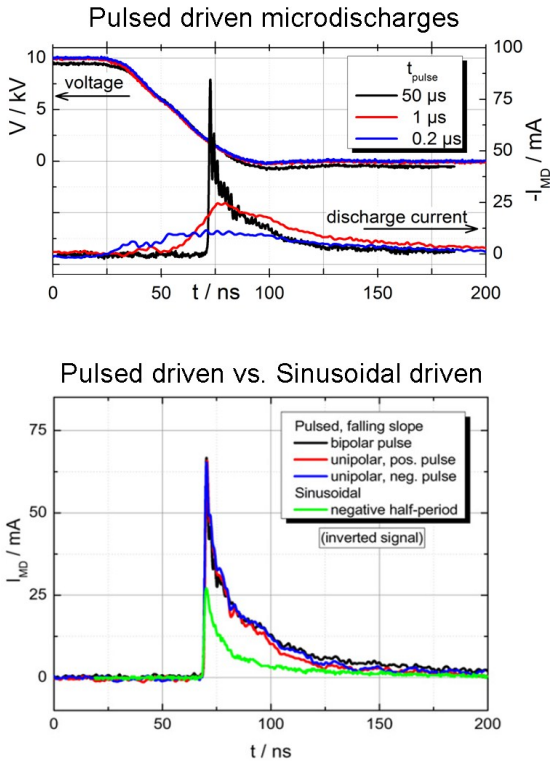

Figure 3. Examples for discharge currents of DBD microdischarges [10].

IV. BREAKDOWN

As mentioned before the investigation of microdischarges requires fast methods with resolution in the sub-ns and sub-mm range. The methods of cross-correlation spectroscopy and streak camera imaging enable the recording of the spatio-temporally resolved development of localized microdischarges [10-12]. Exemplarily a streak photo of a DBD microdischarge in 0.1 vol.% O_2 in N_2 is shown in fig. 4. It has been taken with an optical band filter at 391 nm before the streak camera entrance, i.e. mainly emission of the first negative system of N_2 is recorded. At the conditions being considered the radiating state $N_2^+(B)$ is generated by direct electron collisions with an excitation energy of about 19 eV. Thus the emission can be considered as the development of the high electric field regions.

The microdischarge development can be followed in this figure. A cathode directed ionization front or streamer with a maximum velocity of about 2 10^6 m/s is formed. It was shown in previous studies that this positive streamer starts from the anode region after a Townsend pre-phase which can last some hundreds of nanoseconds. The streamer phase is followed by the formation of the bulk plasma. This plasma region is characterized by a much lower electrical field but higher local electron density.

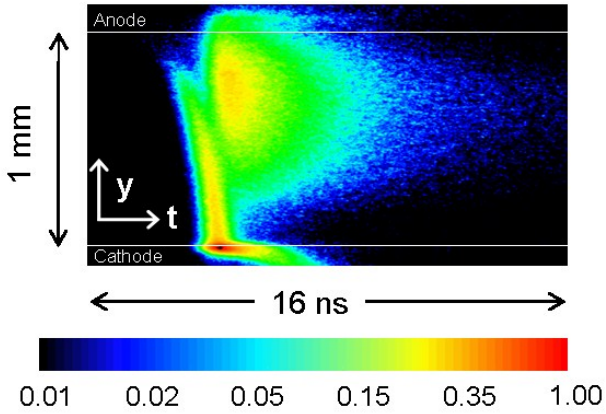

Figure 4. Filtered streak photo (wavelength: 391 nm)

The same principal microdischarge development can be investigated for pulsed and sinusoidal driven DBDs. In particular the microdischarge development in the rising slope is the same for all pulse widths. In contrast the volume breakdown in the microdischarge in the falling slope can be quite different if the switching time t_{pulse} is below the gap crossing time of positive ions [12]. As an example the microdischarge development for t_{pulse} = 1 µs is shown in fig. 5.

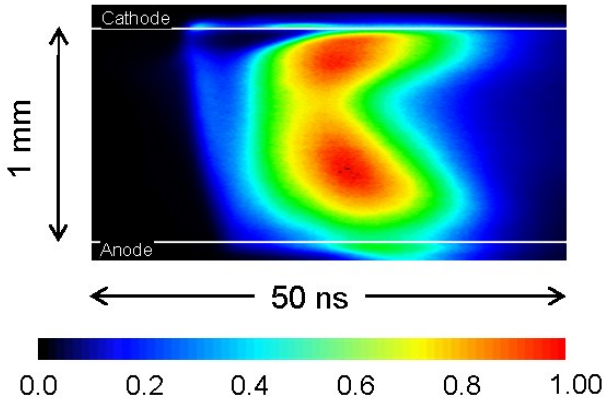

Figure 5. Streak photo for a microdischarge in the falling slope with t_{pulse} = 1 µs [12].

In correlation with the results of the electrical characterization the spatio-temporally resolved development changes significantly. A first emission (in fact a short positive streamer, see) is observed close to the cathode followed by an anode directed streamer and the much brighter bulk plasma with two local maxima close to the electrodes. The streamer velocities are in the order of 10^5 m/s and thus slightly lower than in the usual case without volume pre-ionization. Obviously the pre-ionization depresses the streamer formation which has consequences for the overall microdischarge development and the plasma parameters.

V. SUMMARY

A general overview on the necessary aspects of research regarding the application of Dielectric Barrier Discharges for pollutant degradation in exhaust gases has been given. The importance and possibilities of electrical characterization and the investigation of single microdischarges has been discussed. It has been demonstrated that the dissipation of energy into the DBD microdischarges can be controlled by the shape of the high voltage waveform which is correlated with a manipulation of the streamer-based gap breakdown. In future investigation we plan to discover the consequences of these findings for the plasma chemical conversion. This is motivated by the fact that the plasma properties and parameters which determine the efficacy and selectivity of plasma chemistry can change significantly.

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